Growth and characterization of InGaN/GaN nanoscale heterostructures

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GaN and related compounds are promising materials for light sources fabrication in the visible spectral range. Blue, green and yellow light emitting diodes (LEDs) and CW blue laser diodes (LDs) have been fabricated utilising InGaN single quantum well (SQW) or InGaN/GaN multi quantum well (MQW) structures as active region [1, 2]. In spite of the remarkable progress in the device development, the further improvement in the LEDs and LDs efficiency and reliability and develop of new type of these devices is connected with understanding and detailed studying of both peculiarities of InGaN growth and the radiate recombination process in this material.

In this report, the growth of InGaN/GaN quantum size heterostructures by metalorganic chemical vapour deposition (MOCVD) and its optical characterisation are presented. The structures under studying consisted of InGaN -based active region embedded between GaN layers. These structures were grown on c–plane sapphire in a conventional reduced pressure MOCVD growth machine redesigned for InGaN/GaN growth with a horizontal flow quartz reactor and inductively heated AlN coated graphite susceptor. Ammonia, trimethylindium (TMI), trimethylgallium (TMG) and thrimethylaluminum (TMA) were applied as component precursors. Purified hydrogen and argon were used as carrier gases. Argon was used as a bubbling gas for TMI while hydrogen was used as a bubbling gas for TMG and TMA.

The AlN coating of the susceptor appear to promote the catalytic ammonia decomposition during the low temperature nucleation layer deposition and prevents the destruction of graphite in ammonia ambient [4].

After the substrate heat treatment at $990°C$ in hydrogen ambient, the temperature was reduced and a 30 nm thick Al-containing GaN nucleation layer was deposited at $530°C$ [3]. Then, the substrate temperature was raised to $1050°C$ to grow a 2.5 μm thick GaN layer. During the GaN growth the ammonia and TMG flows were 2.5 sl/min and 36 μmol/min, respectively. The growth rate was 36 nm/min. During these stages the reactor pressure was kept at 200 mbar and hydrogen was used as a carrier gas.

After the GaN growth, the carrier gas was switched from hydrogen to argon, the ammonia flow was increased to 4.5 sl/mim, the TMG flow was ramped down and the reactor pressure was increased to 600 mbar to grow InGaN layers. In accordance with our preliminary experiments, the raise of the reactor pressure during the InGaN growth leads to improvement of material quality and to rise of the indium incorporation efficiency. During switching growth mode from GaN to InGaN and back, the growth process was not interrupted. As in the case of GaN growth, the growth rate of InGaN was found to be proportional to the TMG flow and varied in our experiments in the range of 2.5–6 nm/min.

The structures were characterised by X-ray diffraction (XRD), and photoluminescence (PL) study. Thickness of InGaN layers was estimated on the basis of scanning electron microscope calibration of the growth rate of thick InGaN layers and InGaN/GaN multilayer structures sandwiched between GaN layers. XRD measurements were also employed to determine the period of InGaN/GaN multilayer structures.
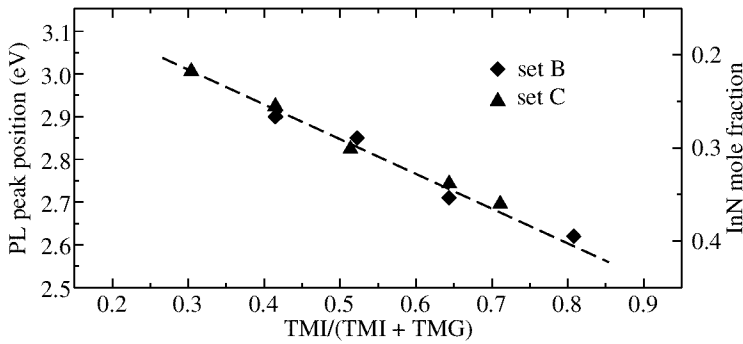
Fig. 1. 77 K PL peak position versus TMI/(TMI+TMG) mole flow ratio for samples from sets B and C.

Three sets of samples with different kinds of InGaN–based active region were prepared. These sets were denoted as A, B and C. The active region of samples of group A consisted of the single thin InGaN layer. During this active layer growth the substrate temperature was reduced to 730°C and then TMI was introduced into the reactor. After the growth of 3 nm of InGaN the substrate temperature was ramped to 1050°C to grow 0.1 μm tick GaN cap layer in hydrogen ambient at pressure of 200 mbar. The TMI flow was switched off from the reactor at the substrate temperature above 800°C.

A strong spatial fluctuations of the InGaN PL peak position from 420 to 500 nm was observed in PL spectra of this samples taken at 77 K. We suppose that this phenomenon is due to nonuniform In content and strain distribution in these lattice-mismatched structures. At the other hand, this effect was not observed for the similar structures with thicker (> 5 nm) InGaN insertions.

The active region of samples of group B consisted of two InGaN layers. First, the substrate temperature was reduced to 800°C to grow 25 nm thick InGaN intermediate layer. Then, keeping all gas flows constant, the temperature was reduced to 730°C and growth of 3 nm InGaN layer was performed. In accordance with our HRD and PL calibrations In content in the intermediate layer was approximately two times lower than in the thin InGaN layer grown at low temperature. The procedure of GaN capping layer growth was the same as in the case of samples of A group.

In the PL spectra of this set of samples no spatial variation of peak position could be observed. Moreover, the PL intensity from InGaN layer was approximately 20 times higher than in the case of A set of samples. The similar effect was previously observed by S. Keller et al. [5]. The peak position of PL from InGaN layer was determined by the TMI/(TMG+TMI) mole flow ratio as shown in Fig. 1. This ratio was varied both by TMI and TMG flows change.

The composition of the ternary alloys $In_xGa_{1-x}N$ was calculated using a standard expression: $E_g(x) = x \times E_{g,InN} + (1 - x) \times E_{g,GaN} - x(1 - x) \times b$, where x represents InN molar fraction, $E_g(x)$ represents the peak of the PL emission of $In_xGa_{1-x}N$ alloy, $E_{g,InN}$ and $E_{g,GaN}$ represent the band-gap energies for InN (1.95 eV) and GaN (3.49 eV) respectively, and b is a bowing parameter ($b = 1.0$ eV) [1] (see Fig. 1). The indium content in the InGaN layers was found to be independent of growth rate in the experimental range.

The active region of samples of set C was similar to the one of samples of set B excepting that it contained several thin InGaN layers with high In content. After the growth of the intermediate InGaN layer the substrate temperature was cycled from 730°C to 860°C keeping all gas flows constant. We grew a number of 5 and 12 period structures. The
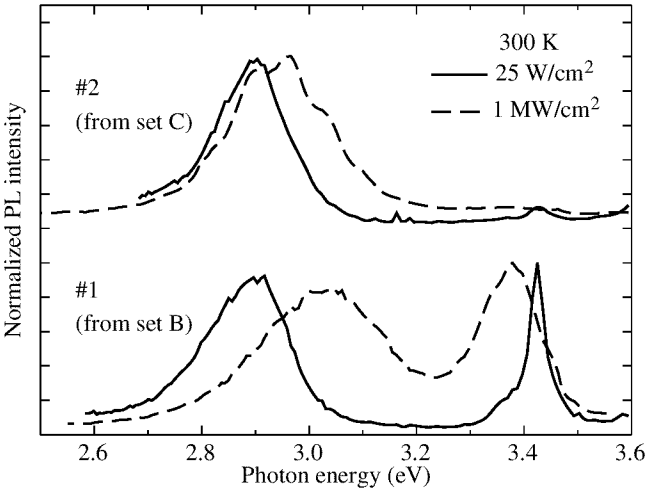
Fig. 2. Room temperature PL spectra at low and high excitation density for samples from sets B and C grown under the same TMI/(TMI + TMG) mole flow ratio of 0.415.
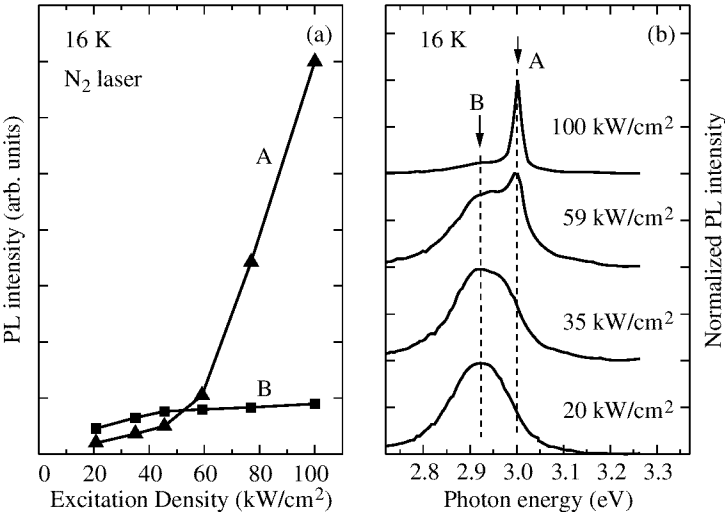
Fig. 3. Photopumped lasing at 16K in sample with cleaved facet mirrors: photoluminescense intensity *versus* excitation density (*a*) and PL spectra (*b*) taken at different excitation densities.

procedure of GaN capping layer growth was the same as in the case of type A samples. The main idea of this growth process was to create a multilayer structure only by growth temperature variation while being other growth conditions unchangeable. Here, the low temperature is needed for increase the indium incorporation in the InGaN layers whereas the high temperature is needed for growth the GaN barrier layers. Indium incorporation in InGaN layer is known to be decreased with the growth temperature increase [2]. The period of the superlattices (SLs) revealed from XRD was 12 nm. This value was in a good agreement with one estimated on the basis of TMG flow. The average In content in the SL from XRD measurements was approximately equal to the In content in the intermediate layers grown at 800°C.

The PL spectra of these samples under low excitation level (He-Cd laser, 25 W/cm^2) were identical to spectra of the samples with single InGaN layer (set B) grown with the same TMI/(TMI+TMG) mole flow ratio (see Fig. 1). In contrast, under high level of excitation (N$_2$ laser, up to 1 MW/cm^2) the broadening of the PL spectra and the blue shift was lower for samples from set C than for samples from set B (Fig. 2). Moreover, samples with 12 period superlattices grown under moderate TMI/(TMI+TMG) mole flow ratio (0.4–0.5) demonstrated lasing under optical pumping in both vertical and horizontal directions (see Fig. 3) [6] at temperatures up to 200 K.

In conclusion, we show that intensity of InGaN—related photoluminescence increased more than an order of magnitude for structures with intermediate InGaN layer. The growth of the InGaN/GaN SL structures was realized by periodically alteration of the growth temperature in the range between 730°C and 860°C. Lasing under conditions of optical pumping was obtained both in directions perpendicular and parallel to sample surface.

References

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